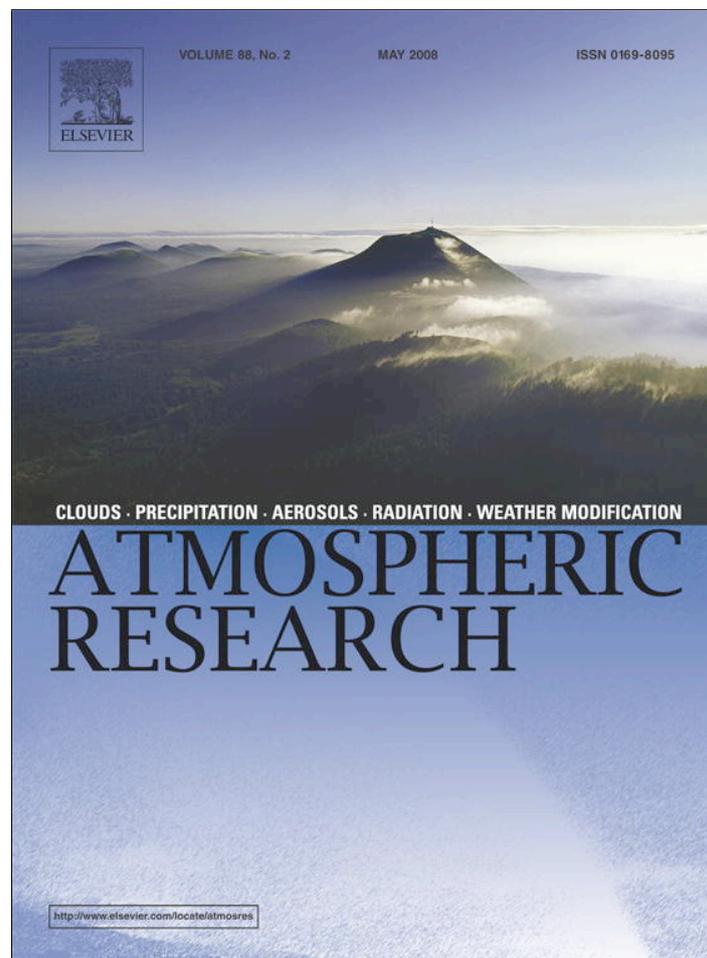


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## Aerosol trends over China, 1980–2000<sup>☆</sup>

David G. Streets<sup>a,\*</sup>, Carolyne Yu<sup>a</sup>, Ye Wu<sup>b,1</sup>, Mian Chin<sup>c</sup>, Zongci Zhao<sup>d</sup>,  
Tadahiro Hayasaka<sup>e</sup>, Guangyu Shi<sup>f</sup>

<sup>a</sup> *Decision and Information Sciences Division, Argonne National Laboratory, Argonne, Illinois 60439, USA*

<sup>b</sup> *Energy Systems Division, Argonne National Laboratory, Argonne, Illinois 60439, USA*

<sup>c</sup> *NASA Goddard Space Flight Center, Greenbelt, Maryland 20771, USA*

<sup>d</sup> *National Climate Center, China Meteorological Administration, Beijing 100871, PR China*

<sup>e</sup> *Research Institute for Humanity and Nature, Kita-ku, Kyoto 603-8047, Japan*

<sup>f</sup> *Institute of Atmospheric Physics, Chinese Academy of Sciences, Beijing 100029, PR China*

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### Abstract

Annual emission trends of sulfur dioxide, black carbon, and organic carbon are presented for East Asia for the period 1980–2000. Emissions of sulfur dioxide peaked in about 1996, and emissions of the carbonaceous aerosols peaked in about 1994–1995, due to a variety of economic, environmental, and social forces. These emissions are converted to their contributions to aerosol optical depth (AOD) over East Asia, using regional results from the GOCART global chemical transport model. We calculate that, on average, AOD over China rose from a value of 0.25 in 1980, peaked at a value of about 0.305 in 1995–1996, and then decreased to about 0.29 in 2000. This trend is consistent with surface shortwave irradiance measurements at 52 weather stations in China, as well as with other radiation-related trends. It may also be consistent with a rise in mean surface temperatures in China starting about the middle of the 1990s.

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*Keywords:* Aerosols; China; Solar radiation; Climate change

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### 1. Introduction

Weather records in China for the second half of the 20th century have revealed fundamental changes in the spatial and temporal patterns of temperature, rainfall, cloud cover, solar radiation, and related variables, which may be partly or entirely explained by changes in aerosol concentrations. Wang and Gong (2000) presented a very long temperature trend from 1880–2000, stressing increasing temperatures in recent decades and identifying 1998 as the warmest year in the record. Yu et al. (2001) analyzed temperature records for 1951–1994 in

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\* Corresponding author. Tel.: +1 630 252 3448; fax: +1 630 252 5217.

E-mail address: dstreets@anl.gov (D.G. Streets).

<sup>1</sup> Now at Department of Environmental Science and Engineering, Tsinghua University, Beijing 100084, PR China.

eastern China, finding a slight warming trend that they tentatively attributed to increasing concentrations of aerosols during winter and spring. Zhai and Pan (2003) reported trends in temperature extremes in China, 1951–1999, finding a decreasing trend in the number of hot days, frost days, cool days, and cool nights. He et al. (2005) analyzed temperature records from 1951–2002 and found the strongest warming trend at higher latitudes during winter. Gong et al. (2006a) were even able to surmise that diurnal temperature variations in China were associated with human-caused fluctuations in aerosol production during weekday and weekend periods. Wang and Zhou (2005) observed significant trends in extreme and mean precipitation events during the period 1961–2001. They found that annual mean precipitation increased significantly in southwest, northwest, and east China, and decreased significantly in central, north, and northeast China; extreme precipitation events in the Yangtze River basin increased dramatically in the summertime. Finally, Zhao et al. (2006) suggested a possible feedback between increasing aerosols over central China and changes in precipitation.

With regard to cloud cover, Kaiser (1998, 2000) found that observational data for the period 1951–1994 showed a decreasing trend over much of China and concluded that the increasing temperature trend could not be explained by increasing cloud cover. In a subsequent analysis of sunshine duration records for China, Kaiser and Qian (2002) found a significant decrease, especially over eastern China, which was attributed to large increases in anthropogenic aerosol loading. Qian et al. (2006) extended the cloud-cover record to 2001, confirming the decreasing trend in cloud cover and showing also a decreasing trend in solar irradiance, based on data in Liu et al. (2004); however, they indicated a possible reversal to a slightly increasing trend in solar irradiance since “about 1990.” Che et al. (2005) analyzed forty years of solar radiation data in China, 1961–2000, finding a long-term decline in direct radiation, clearness index, and possible sunshine duration, but an increase in diffuse radiation; they also cited evidence that conditions improved in the last decade. Kawamoto et al. (2006) demonstrated a correlation between aerosol concentration and cloud properties over East Asia that was consistent with the Twomey effect.

A probable explanation for all of these effects, often explicitly offered in the studies, is an increase in aerosol build-up over China, associated with increasing levels of sulfate, carbonaceous, and other aerosols resulting from China's rapid economic development and growing coal use since the beginning of industrialization. How-

ever, these studies mostly concern themselves with the period from the 1950s to the 1990s, when anthropogenic emissions were undoubtedly on the rise. This paper presents estimates of annual emissions in East Asia from 1980–2000 and shows that aerosol concentrations over China may have peaked in the early to mid-1990s and begun to decrease thereafter. This is shown to be consistent with the observed surface solar radiation trend.

We have previously analyzed trends in global average aerosol optical depth (AOD) and correlated them with trends in emissions of sulfur dioxide (SO<sub>2</sub>), black carbon (BC), and organic carbon (OC) in 17 world regions (Streets et al., 2006). We demonstrated that the global average AOD trend was consistent with the global average trend in solar radiation reaching the Earth's surface, and we postulated that this was the explanation for the so-called “dimming/brightening transition” that shows up in surface radiation data in the late 1980s. Qualitatively, the conclusions also seem to hold for a variety of world regions, and we are now able to test the relationship for one of the most important of those regions, China.

## 2. Methodology and results

In previous work, we presented estimates of China's emissions of various atmospheric pollutants for various time periods (Streets et al., 2000, 2001a,b, 2003; Streets and Aunan, 2005). Conclusions from those previous studies that are relevant to this present work are that (a) SO<sub>2</sub> emissions in China peaked in 1996 (Streets et al., 2000); (b) CO<sub>2</sub> emissions in China during the period 1990–2000 also showed a peak in 1996 (Streets et al., 2001b); and (c) BC emissions in China peaked in 1994–1995 (Streets and Aunan, 2005). This paper presents new estimates of annual aerosol precursor emission trends in China during the period 1980–2000—driven by the same set of energy, fuel-use, and human activity parameters, and incorporating time-dependent trends in emission rates.

In other related work (Streets et al., 2001a, 2004; Bond et al., 2004), we reported the development of detailed inventories of primary carbonaceous aerosol emissions (BC and OC) for Asia and the world. In particular, a detailed global inventory of primary BC and OC emissions was reported for the year 1996 (Bond et al., 2004). We have now extended the 1996 data for East Asia to an annual trend for the period 1980–2000 and adapted the model to estimate annual SO<sub>2</sub> emissions over the same period. We use annual fuel-use trends for East Asia (Streets and Aunan, 2005), processed into 112 technology/fuel combinations (Streets et al., 2004). Incorporated into the emission calculations are time-

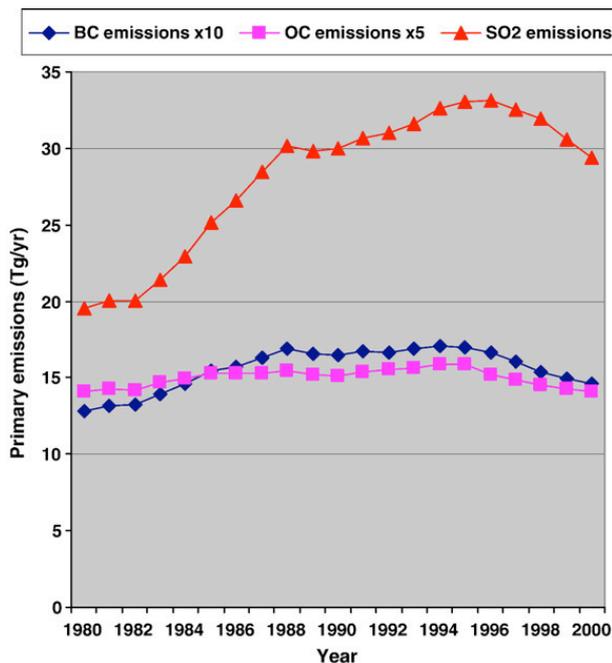


Fig. 1. Primary aerosol emissions in East Asia, 1980–2000, with SO<sub>2</sub> as a proxy for sulfate.

dependent trends in technology penetration, emission controls, and coal sulfur content.

Fig. 1 shows calculated annual trends in SO<sub>2</sub>, BC, and OC emissions during the period 1980–2000. There was a sharp increase in emissions until the mid-1990s that was halted by a variety of economic, environmental, and social forces. The decline in SO<sub>2</sub> emissions after 1996 is attributed to a slowdown in economic growth, a decline in coal use in the residential sector and parts of the industrial sector, and a reduction in the average sulfur content of coal burned (Streets et al., 2000, 2001b). The decline in primary carbonaceous aerosol emissions is attributed to a decrease in the use of residential coal and biofuel caused by a combination of environmental pressure to reduce particulate emissions in cities and measures to foster social development in rural China (Streets and Aunan, 2005). This paper does not address trends in NO<sub>x</sub> emissions and nitrate aerosol production; however, growth in NO<sub>x</sub> emissions in China may also have been arrested in the late 1990s (Hao et al., 2002), though satellite trends (Richter et al., 2005) reveal the beginnings of a dramatic increase in NO<sub>x</sub> emissions in 2000–2001.

For the purposes of comparing the effects of these primary emission changes on the East Asian aerosol burden and subsequent changes in aerosol optical depth (AOD), we have combined all of the major aerosol types in Fig. 2. Here we use GOCART global chemical transport model results from Chin et al. (2002), updated

to GOCART c3.1 simulations for 2000 (Chin et al., 2004), where we have extracted results for East Asia from the global simulations. We use the GOCART model simulations of emissions, mass loading, and AOD for the year 2000 to establish their interrelationships; we then assume that AOD is linearly related to emissions on an annual average over the regional domain of China. Table 1 summarizes the model assumptions and results. We estimate the AOD trend for 1980 to 2000 based on the time-varying aerosol contributions from SO<sub>2</sub> emissions in East Asia (a range of 9.8–16.6 TgS yr<sup>-1</sup>) and BC emissions (range of 1.29–1.71 TgC yr<sup>-1</sup>). Even though we have calculated the trend in primary OC emissions (Fig. 1), we do not know the trend in secondary organic aerosol production, and for that reason we keep the OC contribution to global aerosols constant at the standard model input value 7.47 TgC yr<sup>-1</sup>.

For the purpose of this study, we assume that there are no systematic trends in the source strengths of mineral dust and sea salt over the 20-year period, though we recognize that there can be considerable inter-annual variability—of dust, at least. Mukai et al. (2004) and Gong et al. (2006b) discussed trends in observed and modeled mineral dust over China, using the metric of number of days with dust events. Although there seems to be some evidence for a decreasing trend during this time period, the authors are cautious about their

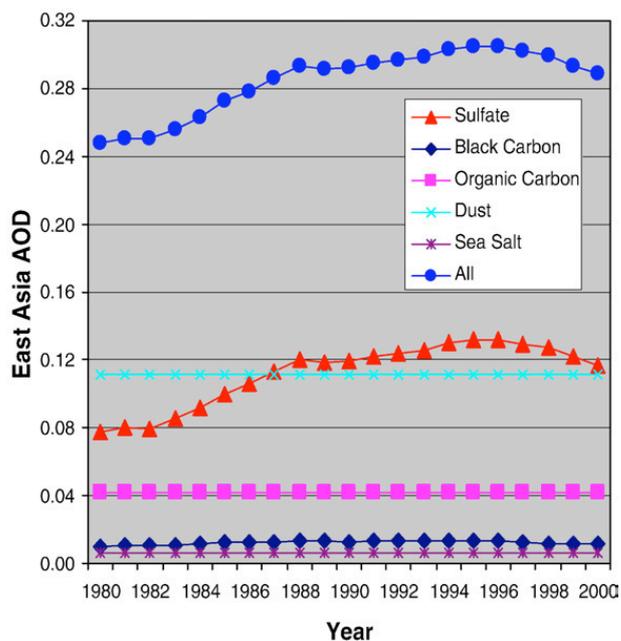


Fig. 2. Trends in East Asia average aerosol optical depth (AOD), 1980–2000. OC, dust, and sea salt are included as constant values over this time period, solely for illustration of their relative contributions to the total; we recognize that they have a natural inter-annual variability.

Table 1  
Emissions, mass burdens, and aerosol optical depths

East Asia					Global	
Aerosol type	Emissions (Tg yr <sup>-1</sup> ) <sup>1</sup>	Atmospheric burden (Tg yr <sup>-1</sup> ) <sup>2</sup>	Aerosol optical depth (at 550 nm) <sup>2</sup>	Aerosol optical depth (%)	Aerosol optical depth (at 550 nm) <sup>2</sup>	Aerosol optical depth (%)
Sulfate <sup>3</sup>	9.8–16.6	0.0511	0.0777–0.132	32–43	0.0290–0.0323	28–30
BC <sup>3</sup>	1.29–1.71	0.0221	0.0100–0.0132	4.0–4.3	0.0025–0.0027	2.4–2.5
OC	7.47	0.0662	0.0420	14–17	0.0143	13–14
Dust			0.111	37–45	0.0342	31–33
Sea salt			0.0066	2.2–2.7	0.0254	23–24
All			0.248–0.305	100	0.105–0.109	100

<sup>1</sup> Source strengths for OC are taken from the GOCART model for the year 2000 (Chin et al., 2004). Sulfate source strength is quantified as primary emissions of SO<sub>2</sub> in TgS.

<sup>2</sup> Global average atmospheric burden and aerosol optical depth are converted from the source strength based on GOCART simulations (Chin et al., 2002, 2004).

<sup>3</sup> Ranges shown in the data for sulfate and BC represent the range of values in the 20-year trend, min–max.

conclusions; and, in any case, the number of major dust event days is so small (generally less than ten per year) that any inter-annual differences will not materially affect the annual average trends presented here. Fig. 2 shows that AOD over China during this time period is considerable, ranging between 0.24 and 0.31, as compared with the global average value of about 0.1. Dust and sulfate dominate the AOD for East Asia, with lesser contributions from OC, BC, and sea salt. The trend of all species combined suggests that AOD rose from a value of about 0.25 in 1980, the beginning of our analysis period, through a minor peak in 1988, to a peak value of 0.305 in 1996, declining thereafter to about 0.29 by 2000. The trend in sulfate aerosol is the major determinant of the AOD trend.

These emission trends and estimated AOD trends are consistent with a number of observations about possible reversal in the 1990s of long-term trends in weather variables. To investigate this possibility, we processed annual mean measurements of surface shortwave irradiance data from 52 stations in China between 1960 and 2000 (Hayasaka et al., 2006). The solar radiation data are from the Meteorological Information Center, which is part of the China Meteorological Administration. A set of quality assessment algorithms has been used to test the quality of daily solar global, direct and diffuse radiation measurements taken at all 122 observatories in China during 1957 to 2000 (Shi et al., in press). The QA algorithms include a physical threshold test, a global radiation sunshine duration test, and a standard deviation test applied to time series of annually-averaged solar global radiation. Although the quality assessment process had significant effects on the data from individual stations and/or time periods, it did not affect the long-term trends in the data. We use data

from a subset of 52 stations that passed the quality assessment process.

The average of radiation trends over all 52 stations is shown in Fig. 3a. We find that the data can be represented by a cubic equation curve obtained by least-squares fitting. The  $R^2$  coefficient of determination is 0.79, and the standard deviations are shown on Fig. 3a. A minimum in the fitted curve is revealed at ~1991. The features of the national trend are nicely exemplified by Beijing (Fig. 3b), the city for which industrial pollution mitigation measures have been most strongly enforced in recent years. The minimum for Beijing occurs a little later, at ~1994. Fig. 4 shows the shortwave radiation trends for each of the individual stations in China during the period 1971–2000, as well as their locations. It is clear from this figure that the greatest decreases in radiation reaching the Earth's surface during this period are in the central and central coastal regions of China where coal-based industrial development was highest. Western China and some southern and north-eastern regions show much less decrease, associated with lack of development or situations with favorable meteorology.

A similar minimum is visible in the pan evaporation and solar irradiance trend data for China reported by Liu et al. (2004) and Qian et al. (2006). Inspection of their trend curves suggests a minimum in the pan evaporation trend at ~1993 and in the solar irradiance data at ~1989. The data of Che et al. (2005) reveal minima in daily clearness index at ~1989 and in relative sunshine duration at ~1993. The long-term AOD data reported by Luo et al. (2001) over China cover the period 1960–1990 and show a steeply increasing trend to ~1982, with a moderation thereafter. The maximum in our AOD trend occurs somewhat later than the

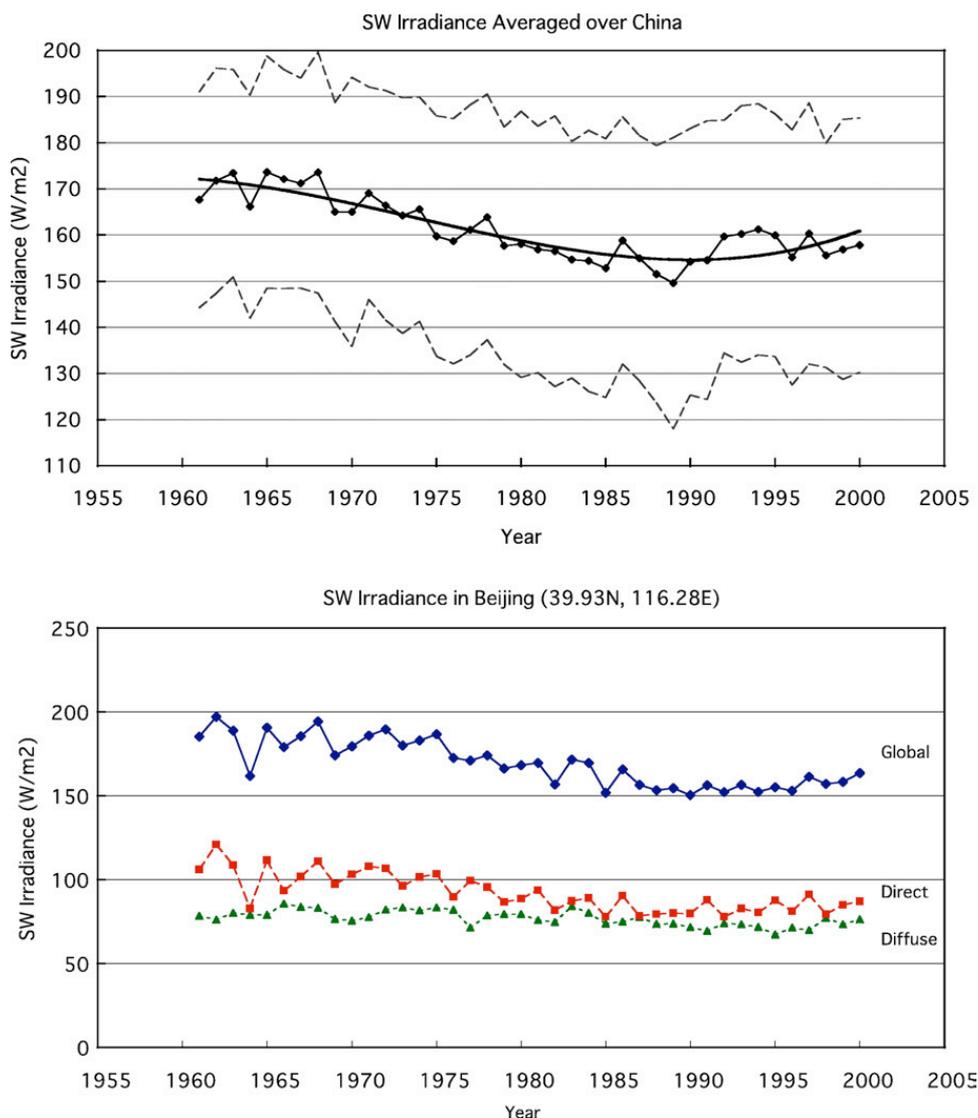


Fig. 3. Shortwave irradiance measurements ( $\text{W m}^{-2}$ ) in China, 1960–2000, averaged for 52 monitoring stations (3a, upper) and for Beijing (3b, lower).

minima in the radiation data, which may be due to our inability at present to quantify the time-varying contribution of OC to AOD over China; primary emissions of OC are somewhat similar to emissions of BC, though muted because of the larger contribution to OC from open biomass burning, which does not change in our emission trends. In future work we will investigate the trend in secondary organic aerosol production in East Asia, which requires determinations of the trends in primary VOC emissions from both anthropogenic and biogenic sources—neither of which have thus far been attempted. In other future work we plan to simulate the AOD trend from 1980 to the present with full annual runs of the global chemistry transport model, in which the inter-annual variation of anthropogenic, biomass burning, mineral dust, and sea-salt emissions will be

taken into account, such that the aerosol-induced changes of surface radiation can be more accurately estimated for the entire Earth's surface.

Our hypothesis may also be consistent with recent temperature trends in China, though this requires further work to disentangle the many contributing factors to climate modification. The long-term temperature trend exhibits a period of cooling due to aerosols over China that began in about 1950 (Zhao et al., 2005; Wang and Gong, 2000). Analysis of temperature anomalies for the period 1980–2000 (Fig. 5) appears to show an increase that could be consistent with a decline in average aerosol concentrations (Zhao et al., 2005). Unfortunately, there is no clear onset that can be discerned in the record: the observations (solid black line) do show some indications of a greater increase in the 1990s, but the inter-annual

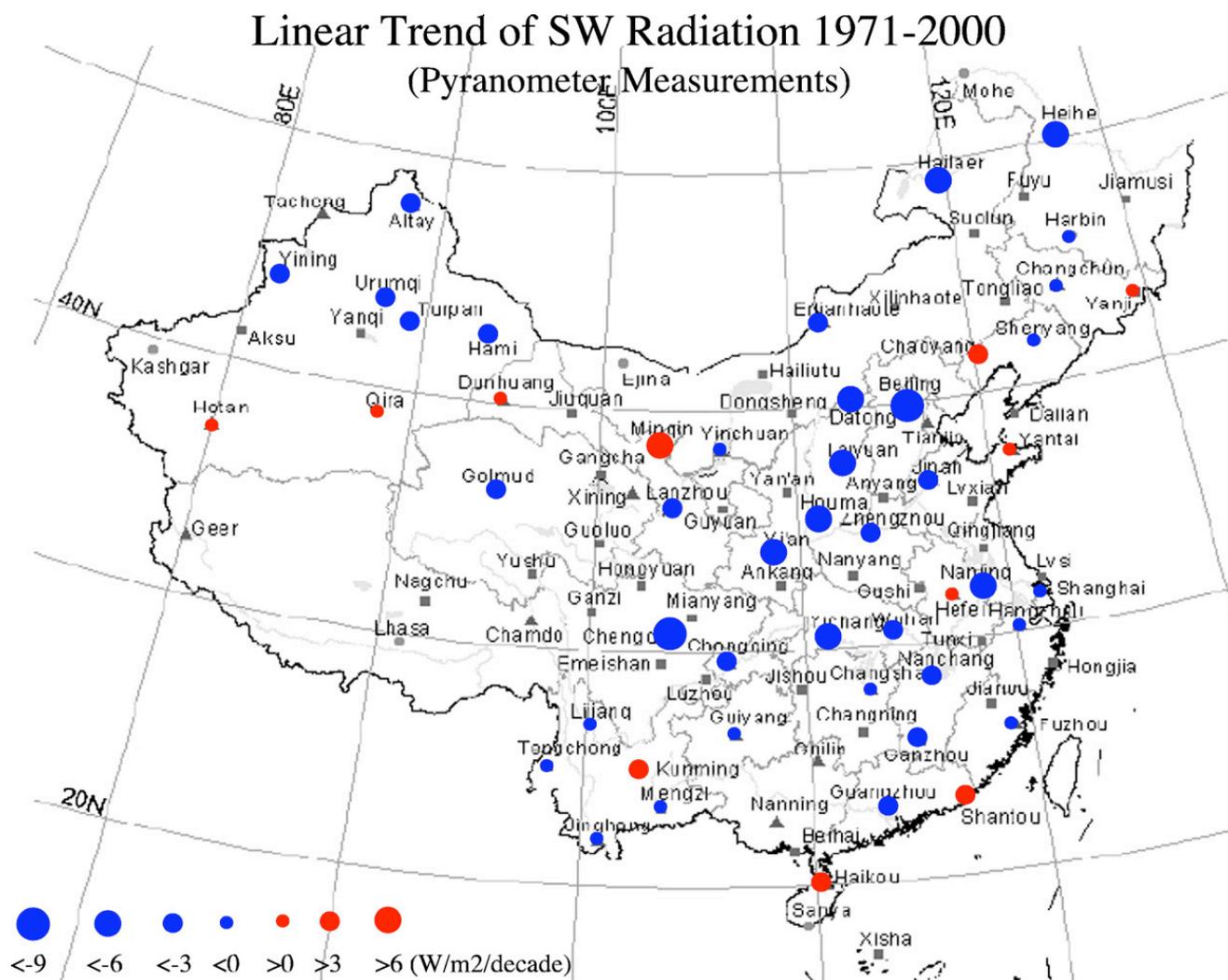


Fig. 4. Changes in shortwave pyranometer measurements at individual stations in China during the period 1971–2000, in  $\text{W m}^{-2} \text{decade}^{-1}$ .

variations are so dramatic as to prevent a definitive interpretation.

Naturally, there are also many other influences on temperature trends, besides man-made aerosols, that confound a simple explanation. These include volcanic eruptions such as El Chichon (1982) and Pinatubo (1991) and the effects of El Nino and La Nina events—not to mention the fact that increasing  $\text{CO}_2$  and other greenhouse-gas emissions may have infused a long-term warming contribution in the past decade or two. Further analysis of meteorological and climatological variables in the 1980s and 1990s is needed to confirm the temperature trends and their causes.

### 3. Discussion

We present annual trends in man-made emissions of aerosol-relevant species in East Asia for the period

1980–2000 and convert them into average AOD trends over China. Our analysis is consistent with other studies in affirming that there was a steady rise in regional aerosol concentrations since the start of industrialization in China in the 1950s that caused a decline in solar radiation and regional cooling (due to sulfate). However, we suggest that emissions peaked in the early to mid-1990s and began to decline thereafter. Improved coal-burning methods in the power generation and industrial sectors, coupled with a transformation away from the burning of solid fuels in the home, were the primary causes of this change.

Analysis of weather variables prior to the mid-1990s at Chinese weather stations reveals a pattern of steady or slowly changing temperature, decreasing solar radiation at the surface, and modified precipitation patterns. These trends are all consistent with a build-up of atmospheric aerosols, in which the natural

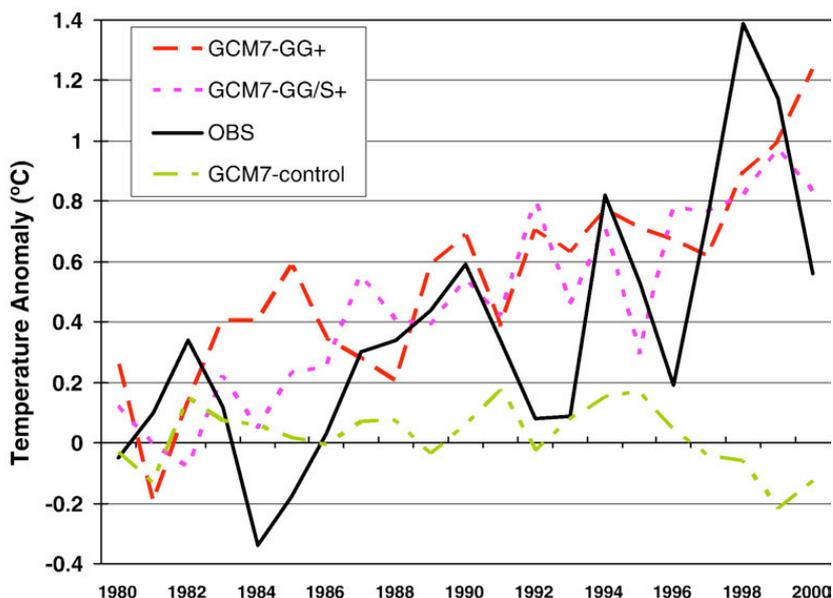


Fig. 5. Evolution of annual mean surface air temperature anomalies over China and East Asia from 1980 to 2000 (relative to 1961–1990): OBS = observations; GCM7-GG+ = the average of seven GCMs with greenhouse-gas emissions increasing; GCM7-GG/S+ = the average of seven GCMs with both greenhouse-gas emissions and sulfate aerosols increasing; GCM7-control = the average of seven GCM control runs [the seven GCMs are CCC-Canada, CCSR/NIES-Japan, CSIRO-Australia, DKRZ-Germany, GFDL-USA, HADL-UK, NCAR-USA].

contributors like dust and sea salt are enhanced by increasing amounts of man-made sulfate, carbonaceous aerosols, and other species. Climate/chemistry modeling has been able to successfully reproduce the effects of sulfate, black carbon, and other kinds of particles over China (Menon et al., 2002; Qian et al., 2003; Huang et al., 2006; Gu et al., 2006; Rangwala et al., 2006). As the state of the art of aerosol modeling progresses, model results are increasingly able to explain the relationships between emissions, aerosol

loadings, and weather and climate factors, and mostly they support the empirical correlations between observed parameters that have been made during the last two decades. This paper suggests that the most recent analyses of observations in China provide the first evidence that the trend of increasing aerosol concentrations, decreasing solar radiation, and corresponding climate effects may have reversed itself in the 1990s, leading to an increase in solar radiation at the surface and possibly a rise in mean surface

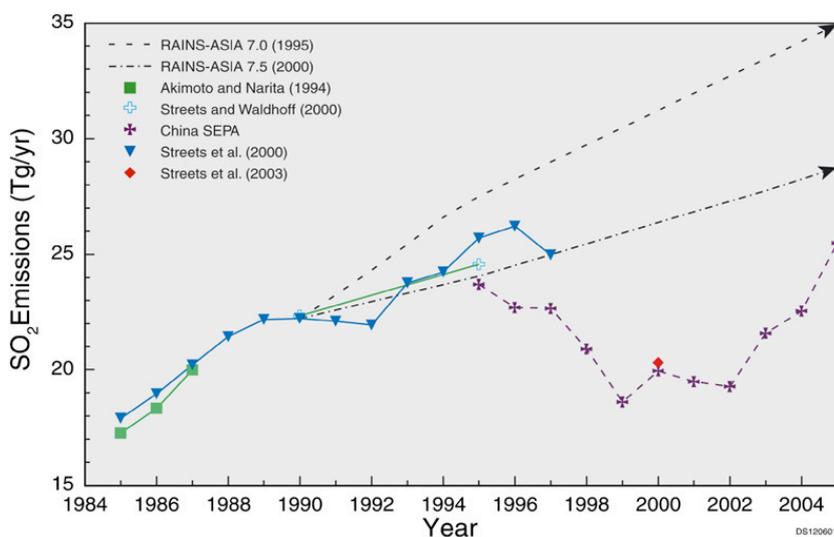


Fig. 6. Trends in SO<sub>2</sub> emissions in China, 1985–2005.

temperatures, in accordance with expectations from our calculations of emission and aerosol trends. For decades, an aerosol haze has helped to shield China from solar radiation, but in the future the country may return to a hotter, drier regime, with all that that implies for agricultural productivity, water availability, etc. (Yang et al., 2005).

Whether the trends of Fig. 1 will continue into the future depends on (a) the extent to which future economic growth is powered by coal and oil; (b) the extent to which increased control of emissions will counteract China's appetite for fossil fuels; and (c) the rate at which social development will displace coal and biofuels from cooking and heating usage in the home. Since 2000, the trend has not been promising. Fig. 6 shows trends in SO<sub>2</sub> emissions from official sources (the China State Environmental Protection Administration (China SEPA)) and other studies (Akimoto and Narita, 1994; Streets and Waldhoff, 2000; Streets et al., 2000, 2003) during the period 1985–2005. The gains that were achieved through declining emissions from 1995–1999 have been lost during the period 2000–2005. Rapidly expanding electricity generation and increasing industrial production—all fueled by coal—have led to a return of SO<sub>2</sub> levels to those of the mid-1990s. This has been acknowledged by SEPA as one of the two most significant environmental failures of the 10th Five-Year Plan. Thus, it will be interesting to see if solar radiation levels were progressively reduced (increased dimming) during the first half of the present decade. SEPA has made a renewed effort to halt this trend during the period of the 11th Five-Year plan, so that by 2010 SO<sub>2</sub> emissions will be capped at 23 Tg/yr. If the planned measures are not successful, the long-term trends predicted five years ago by the RAINS-ASIA model Version 7.5 (Shah et al., 2000), or even ten years ago by the RAINS-ASIA model Version 7.0 (Foell et al., 1995), as shown in Fig. 6, may prove to be not so far wrong after all.

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